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Dielectric properties of polycrystalline $(Ba_{0.60}Sr_{0.40})Ti_{0.8}O_3$

M. Gabryś*

Institute of Physics, Pedagogical University, ul. Podchorążych 2, 30-084 Kraków, Poland

* Corresponding author: E-mail address: mgabrys@ap.krakow.pl

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ABSTRACT

Purpose: The purpose of the work was to determinate the influence of the ferroactive Sr substitutions in sublattice A and the nonstoichiometry in sublattice B on changes of physical properties and the character of phase transition (PT) in pure barium titanate $BaTiO_3$ (BT).

Design/methodology/approach: The polycrystalline samples of $(Ba_{0.60}Sr_{0.40})TiO_3$ and $(Ba_{0.60}Sr_{0.40})Ti_{0.8}O_3$ were obtained by the calcinations method in temperature 1620 K. The dielectric measurements were executed by automatic device (QUATRO KRIO 4.0 with LCR Agilent 4824A meter and BDS 1100 cryostat). The materials were investigated under cooling conditions with speed of 2 K/min and within frequency range from 20 Hz to 1 MHz.

Findings: The dielectrometry was applied to measure complex dielectric permittivity and other dielectric functions of ferroelectric $(Ba_{0.60}Sr_{0.40})TiO_3$ (BST-40) and $(Ba_{0.60}Sr_{0.40})Ti_{0.8}O_3$ (BST-40/0.8). It was affirmed, that 40% substitution of Sr ions as well as 20% deficiency of Ti ions in solid solution reduced temperature and changed the type of phase transformation. The transformation stood strongly diffused. The weak dependence of temperature T_m (peak of electric permittivity ε') from frequency of electric measuring field was observed. It means, that this material should be prescribed to the class of ferroelectrics with diffused phase transformation (DPT). The polar character of this solution was also observed in the paraelectric phase. It is connected with the occurrence of polar clusters in paraelectric phase.

Practical implications: Results can be used to construct the model describing changes in the solid solutions with ferroactive and nonferroactive substitutions in sublattice A or B of the perovskite.

Originality/value: Value of this work relies on the experimental examination of the electric properties of nonstoichiometric BST-40 solid solution. The low value of phase angle in the paraelectric phase was connected with the occurrence of the polar regions.

Keywords: Ferroelectric; Solid solution; Phase transformation; Dielectric properties; Cluster

PROPERTIES

1. Introduction

The solid solution BST-40 is one of the ferroelectric materials of type (A', A'')BO_3. Both components of this solution are ferroelectrics. The pure barium titanate forms 4 structural phases [1, 2]. At high temperatures (in the paraelectric phase) it has a cubic structure. Three phase transitions take place during decrease of temperature to tetragonal phase T (~400 K), then to rhombic phase, O (~300 K) and to rhombohedral phase, R (~210 K). The strontium titanate can be in cubic phase at temperature above 105 K [3].

The aim of the presented work is to determine the influence of ferroelectric component $SrTiO_3$ (ST) and the shortage of Ti ions on the physical properties of BT and also to analyze the change of character and the temperature of phase transitions in this material. Atoms of strontium belong to group of the ferroactive substituentes. The ions Ba and Sr have the similar parameters of external orbitals. Because of that, linear changes of values of physical properties in different Sr – concentration can be expected. Titanium's shortage can cause broadening of phase transition temperature region [4, 5]. This fact can have essential meaning in applications. The analysis of influence of the ferroactive and the nonferroactives ions on structure of crystalline perovskite can explain they role in PT of ferroelectric materials.

Barium titanate is used as capacitor ceramics, piezoelectric transducers, thermistor and chemical sensors [6-8]. Strontium titanate is used in varistor and tunable microwave filters. At very low temperatures, strontium titanate exhibits piezoelectric and superconducting characteristics.

2. Experimental

The polycrystalline samples of $(Ba_{0.60}Sr_{0.40})TiO_3$ and $(Ba_{0.60}Sr_{0.40})Ti_{0.8}O_3$ were obtained by the calcinations method in temperature 1620 K. The dielectric measurements were executed by automatic device (QUATRO KRIO 4.0 with LCR Agilent 4824A meter and BDS 1100 cryostat). The materials were investigated under cooling conditions with speed of 2 K/min and within frequency range from 20 Hz to 1 MHz.

3. Results and discussion

Figures 1-3 present the temperature changes of real component of electric permittivity ε'(T) for pure BT, stoichiometric BST-40 and nonstoichiometric BST-40/0.8. Fig. 1 shows three clear maximums. They involve following phase transitions: C (cubic) – T (tetragonal) – O (orthorhombic) – R (rhombohedral). Fig. 2 shows diffuse phase transitions: C – T and T – O. Irregular decrease of the ε' in the transitions: C – T and the strongly diffused transition T – O (from side of low temperatures) are visible on Fig. 3. The temperature T_m of the C – T transition for all frequencies of measuring field is 273 K. It classifies the BST-40 as a material with DPT. The nonstoichiometry in BST-40/0.8 leads to the increase and the stabilization of values of ε' below the room temperature.

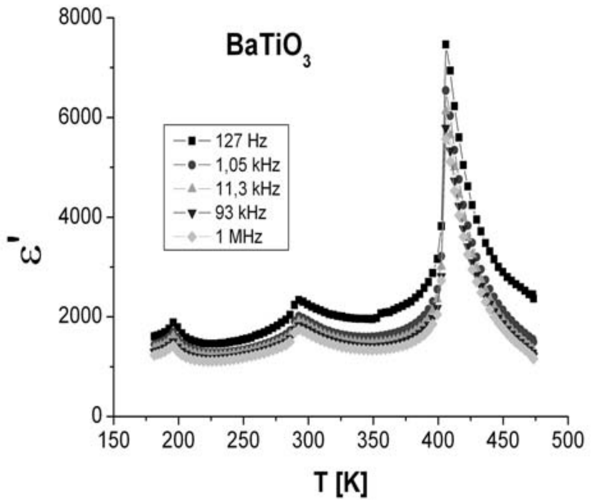

Fig. 1. Temperature dependence of dielectric constant ε' for polycrystalline BT sample

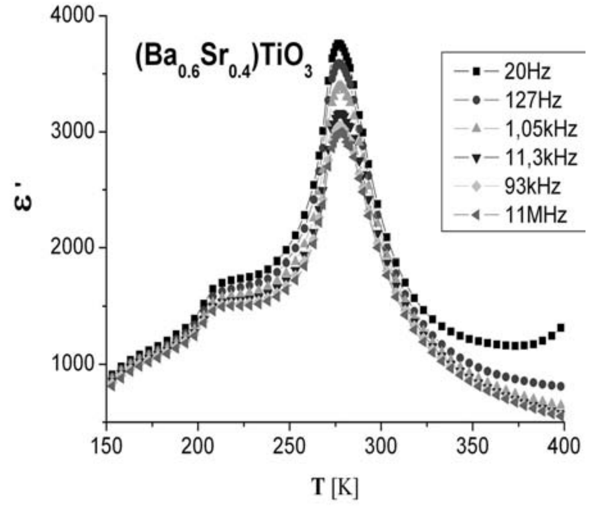

Fig. 2. Temperature dependence of dielectric constant ε' for polycrystalline, stoichiometric BST-40

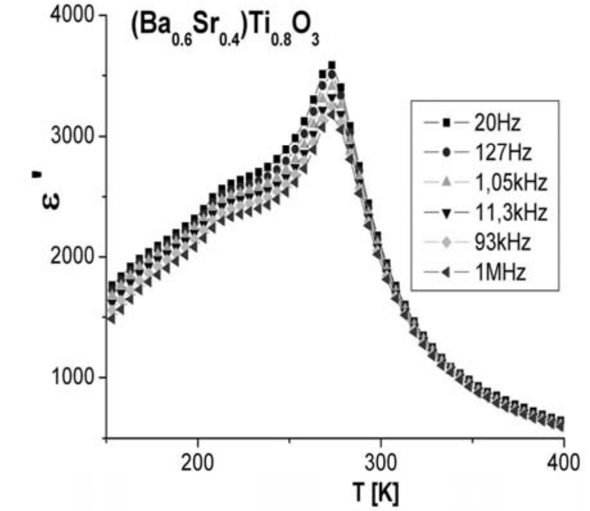

Fig. 3. Temperature dependence of dielectric constant ε' for polycrystalline, nonstoichiometric BST-40/0.8

The following formula describes ferroelectric materials with DPT:

$$\varepsilon^{-1} = \varepsilon_m^{-1} + A(T - T_m)^{\gamma} \tag{1}$$

where ε_m is the maximum value of electric permittivity ε', T_m is the temperature of maximum of electric permittivity ε', A is a constant and γ is an index.

In DPT the value of γ is close to 2. These values were compiled in Figures 4-7, where:

$$y = \varepsilon^{-1} - \varepsilon_m^{-1} \quad (2)$$

$$x = T - T_m \quad (3)$$

Fig. 4 presents dependence of logy = f(logx) for stoichiometric BST-40. Coefficients γ values which characterize a type of PT were determined in Fig. 5.

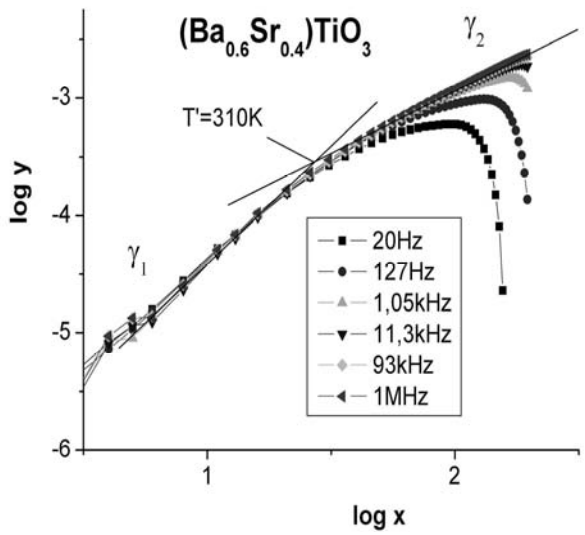

Fig. 4. Dependence logy(logx) for polycrystalline, stoichiometric BST-40

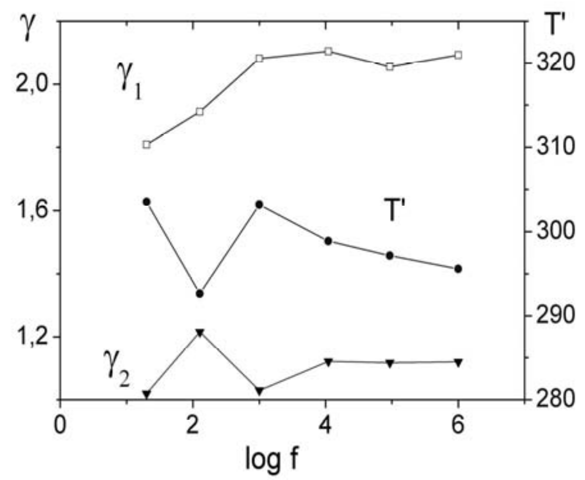

Fig. 5. Dependence $\gamma_1(f)$ and $\gamma_2(f)$ in paraelectric phase for polycrystalline, stoichiometric BST-40

Values $\gamma_1(f)$ and $\gamma_2(f)$ were increasing with growth of frequency f of electric measuring field (Fig. 5). Value $\gamma_1(f)$ was close to 2. This value is typical to DPT. The increase of sample's temperature above T' (Fig. 4, Fig. 6) change the slope of the curve. The γ value jumped from γ_1 to γ_2. The last value is similar to that one of value for the pure BT. The small decrease of temperature T' was also observed. Fig. 6 presents the dependence logy = f(logx) for nonstoichiometric BST-40/0.8. In comparison from stoichiometric sample the linear part of curve for T > T' undergoes the stabilization (is weakly dependent on frequency f).

Fig. 7 presents $\gamma_1(f)$ and $\gamma_2(f)$ as well as the temperature T' in function of frequency of electric measuring field for nonstoichiometric BST-40/0.8. It shows, that values γ_1 and γ_2 grow weakly and temperature T' decreases with frequency of measuring field. Temperatures T' are similar to temperatures in which the curves of dependence of ε'(T) in paraelectric phase are split.

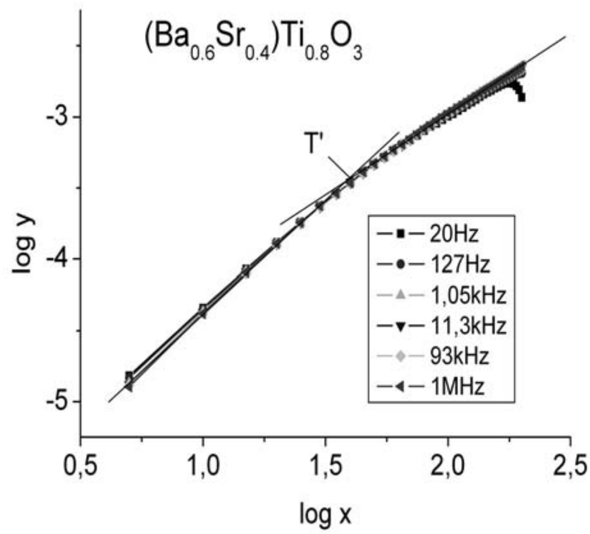

Fig. 6. Dependence logy(logx) for polycrystalline, nonstoichiometric BST-40/0.8

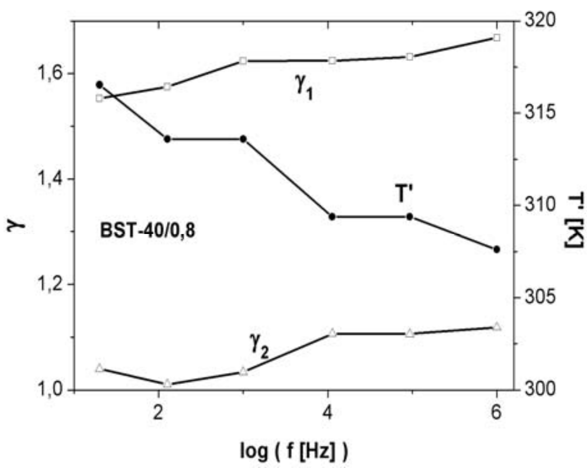

Fig. 7. Dependence $\gamma_1(f)$ and $\gamma_2(f)$ in paraelectric phase for polycrystalline, nonstoichiometric BST-40/0.8

The comparison of coefficients γ_1 and γ_2 as well as the T' leads to conclusion, that their values decreases in nonstoichiometric sample.

Below 470 K values of phase angle lie in compartment from –90 deg to –80 deg. This means, that in observed changes of phase angle with temperature, the polar substructures play decisive part. They create clusters changing with temperature. These clusters are the source of dipolar polarization P_d [9, 10]. This polarization originates from the reaction of electric dipoles short range interactions. The growth of clusters as well as their long range structural and electric collectivization leads to creation of ferroelectric domains. These domains come into being in the area of the paraelectric – ferroelectric PT.

4. Conclusions

The study presents results of dielectric measurements of the BST-40. It was confirmed that the 40% substitution of the ferroactive ions Sr (in sublattice A) lead to strong DPT and to lowering of the temperature of paraelectric – ferroelectric PT. In presented range of temperatures ST is the paraelectric material. However, it does not lead to freezing of cubic structure below the temperature of the paraelectric – ferroelectric PT. This behavior is different as compared to $Pb(Cd_{1/3}Nb_{2/3})O_3$ [11] and $Ba(Ti_{90}Sn_{0.10})O_3$. In the last case, nonferroactive ions Sn make impossible transition of BTS-10 to tetragonal structure.

Nonstoichiometry in sublattice B increases and stabilizes the value of electric permittivity below room temperature. This fact can have essential meaning in technical aplications of this material. The change in concentration of the ferroactive Sr ions can be applied in practice to change of work temperature for electromechanical elements. Polycrystalline BST-40 can be used in the same way as other ferroelectric materials [12-15].

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